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Running title: Electrochemical septic tank treating black water.

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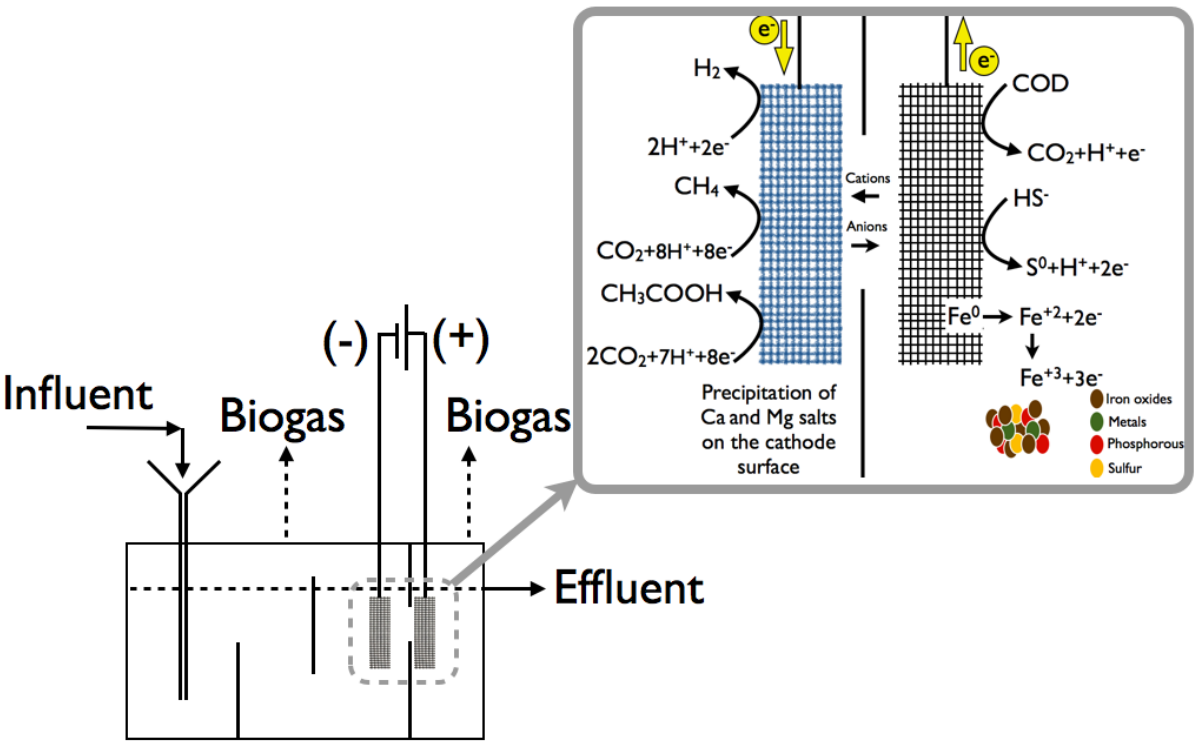
Graphical abstract:



Abstract

Septic tanks are used for the removal of organic particulates in wastewaters by physical accumulation instead of through the biological production of biogas. Improved biogas production in septic tanks is crucial to increase the potential of this system for both energy generation and organic matter removal. In this study, the effect on the biogas production and biogas quality of coupling a 20 L lab-scale septic tank with a microbial electrolysis cell (MEC) was investigated and compared with a standard septic tank. Both reactors were operated at a volumetric organic loading rate of 0.5 gCOD/L d and a hydraulic retention time between 20 and 40 days using black water as an input under mesophilic conditions for a period of 3 months. The MEC-septic tank was operated at an applied voltage of 2.0 ±0.1V and the current experienced ranged from 40 mA (0.9 A/m^2 projected electrode area) to 180 mA (5 A/m^2 projected electrode area). The COD removal was of the order of 85% and the concentration of residual COD was not different between both reactors. Yet, the total phosphorous in the output was on average 39% lower in the MEC-septic tank. Moreover, the biogas production rate in the MEC-septic tank was a factor of 5 higher than in the control reactor and the H_2S concentration in the biogas was a factor of 2.5 lower. The extra electricity supplied to the MEC-septic tank was recovered as extra biogas produced. Overall, it appears that the combination of MEC and a septic tank offers perspectives in terms of lower discharge of phosphorus and H_2S, nutrient recuperation and a more reliable supply of biogas.

Keywords: biogas; energy recovery; electrode; fuel cell; biomethanation.

1. Introduction

Septic tanks are used for decentralized primary treatment of sewage in both developing and industrialized countries. In the United States of America, for instance, approximately 20% of the population relies on septic tanks for the primary treatment of their sewage [1]. The design of septic tanks does not require moving parts and requires little maintenance. It does however require periodical removal of precipitates. Thus, septic tanks can be considered as a relatively robust technology although not inexpensive (i.e. Capex of about €3 000 per family in industrialized countries) [2]. The primary wastewater treatment of conventional septic tanks is limited because the system relies on the capacity of retaining suspended solids by accumulation and sedimentation. Furthermore, most of the dissolved organics and nutrients do not receive significant treatment [3]. In addition, due to anaerobic conditions in the septic tanks, greenhouse gases (GHGs) such as methane (CH_4) can be emitted to the atmosphere instead of being collected. For instance, CH_4 emissions from septic tanks have been measured to be as high as 16 m^3 of CH_4 per inhabitant per year which is a significant source of GHGs [4]. Consequently, the development of approaches to improve biogas production performances of septic tanks is of interest.

Several changes in design have been proposed to improve the performance of COD removal in conventional septic tanks such as the incorporation of aeration and improved anaerobic digestion with or without mixing [5; 6]. The anaerobic process combines the removal of organics, energy recovery and lower sludge production. The combination of an Upflow Anaerobic Sludge Blanket and a septic tank (UASB-septic tank) enhances the contact between the sludge bed and the sewage, increases the removal of suspended solids and allows methanation of dissolved organic compounds [7]. In the context of source separated decentralized water treatment, the UASB-septic tank has been proposed for the treatment of black water as well [6-8]. Black water (mainly faeces and urine) has high concentrations of organic material and nutrients, making it a suitable input for anaerobic digestion. Yet, such a system requires the substitution of a standard septic tank by another reactor. Electrochemical assisted anaerobic digestion has been introduced as a new alternative to improve the anaerobic digestion process [9-11]. The implementation of a microbial electrolysis cell (MEC) coupled with a digester can allow *in situ* production of H_2 and O_2 by applying an electric field. Therefore, an increase of energy content of the biogas can be expected due to a higher H_2 concentration in the biogas and/or a higher production of CH_4 through hydrogenotropic or homoacetogenic pathways [12; 13]. Moreover, it has been shown that direct methanogenesis can occur at electrodes where methanogenic archaea are able to convert electrons, protons and CO_2 into CH_4 [11]. Alternatively, production of micro-aerobic conditions at the anode can increase hydrolysis rates and decrease the concentration of H_2S in the biogas [14-17]. MECs have been coupled to lab-

scale high rate digesters such as UASB reactors treating synthetic wastewater [10; 18; 19]. To our knowledge, MEC systems have not been coupled to reactors with low mixing and complex organic substrates such as septic tanks. Thus, electrochemical assisted anaerobic digestion might offer additional potential to enhance biomethanation while improving biogas quality in decentralized systems. Such information is essential to gain further understanding of a potential new application of electrochemical assisted anaerobic digestion.

In this study, the performance of the anaerobic digestion of black water in a lab-scale MEC-septic tank under mesophilic conditions, at a low organic loading rate and a moderate hydraulic retention time was investigated. An additional lab-scale standard septic tank was operated with the same operational conditions as a control reactor.

2. Material and Methods

2.1 Black water characteristics

Black wastewater was simulated by combining primary sludge from the Breda wastewater treatment plant (Breda, The Netherlands) (30% v/v), pig manure (10% v/v) and tap water (60% v/v) to make up a mixture, in chemical oxygen demand (COD) and total ammonium nitrogen (TAN) basis, of about 10 gCOD/L and 500 mgTAN/L. In practice, the COD in the influent ranged from about 10 to 20 gCOD/L and from 300 to 600 mgTAN/L due to changes in the influent concentration of the primary sludge. Despite these fluctuations, the organic loading rate was kept constant at about 0.5 gCOD/L d creating fluctuations in the hydraulic retention time (HRT). The black wastewater was prepared every 2-3 weeks and was stored at 4° C. The main characteristics of the black wastewater used for the operation of the digesters are given in Table 1.

Table 1. Average characteristics of the influent black water used for the whole experiment.

Parameter	Black water
Total chemical oxygen demand, COD_{total} (g/L)	15.5±3.3
Total volatile fatty acids, VFA_{total} (g/L)	0.4±0.3
Total solids, TS (g/L)	12.0±2.9
Volatile solids, VS (g/L)	8.1±2.1
Total suspended solids, TSS (g/L)	8.3±2.5
Total Kjeldahl nitrogen, TKN (mg/L)	904±152
Total ammonium nitrogen, TAN (mg/L)	499±83
Total phosphorous, P_{total} (mg/L)	324±232
Conductivity (mS/cm)	6.4±0.9
pH	7.9±0.5

2.2 Experimental set-up

2.2.1 Standard septic tank and MEC-septic tank

Both septic tanks were closed rectangular boxes made of Plexiglas frames (Fig. 1). The standard septic tank (control reactor) consisted of a digester of 20 L (working volume) with outer dimensions of 40 × 25 × 25 cm and one biogas output (Fig. 1a). The MEC-septic tank had a total working volume of 24.2 L in two compartments; (i) cathode and (ii) anode compartment (Fig. 1b). The cathode compartment had a working volume of 20 L (40 × 25 × 25 cm) and the anode compartment had a working volume of 4.2 L (10 × 25 × 25 cm). The compartments were separated only in the gas phase allowing an independent collection of the gases from the cathode and anode compartment respectively. For the calculation of organic loading rates (Bv) and the hydraulic retention time (HRT), only the cathode compartment volume was considered because the latter had the same volume in both reactors and the anode reactor volume was proportionally small. Organic loading rates were calculated by multiplying the flow rate of influent (L/d) with the concentration of COD (gCOD/L) divided by reactor volume (20L for both reactors). The biogas production rates were estimated by dividing the amount of biogas production per day (L/d) by the reactor volume (20L for both reactors). The COD mass balance was calculated considering the COD mass input (gCOD) in the reactor and the COD mass output (gCOD in the effluent and gCOD equivalents in the biogas, 1 gCOD = 0.35L CH_4 at STP). Both digesters were inoculated (5 gVSS/L reactor) with granular seed sludge harvested from a full-scale anaerobic digester wastewater and operated at mesophilic conditions (33 ±2 ºC). Biogas production was measured by means of a liquid displacement digital gas meter. The pH, temperature,

COD volatile fatty acids (VFA) and biogas production were monitored and are reported at STP conditions.

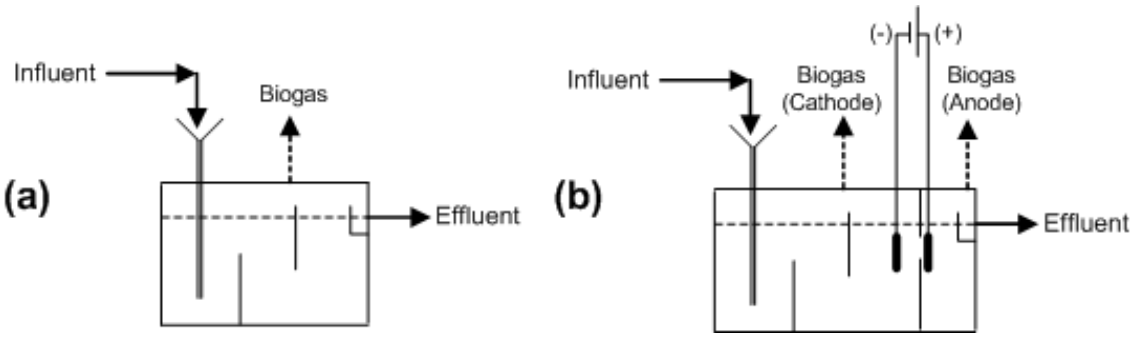


Figure 1. Schematic representation of the lab-scale experimental set-ups. (a) Standard septic tank digester (control reactor) and (b) MEC-septic tank digester.

2.3 Experimental operation

The operation of the MEC-septic tank was divided into two phases: Phase 1 (0–74th day) and Phase 2 (75th–98th day) are considered as the experimental period. The control reactor was operated during the whole experimental period (0–98th day).

2.3.1 Start-up period

Before the experimental period, both reactors had a start-up period. During the latter period of 21 days, both reactors were operated at a maximum organic loading rate (Bv) of 0.51 ±0.01 gCOD/L d with a COD removal efficiency and biogas production rate in the same order for both reactors. Only during the start-up period, the MEC-septic tank was operated with an applied voltage (E_{ap}) of 1.2 ± 0.3 V. The results from the start-up period are not discussed.

2.3.2 Experimental period

Both reactors were fed twice per week in fed-batch mode without pH control in a constant temperature room (30 ±2 °C). The MEC employed two types of stainless steel (AISI 316) mesh electrodes without membrane in two phases (Fig. 1). Rectangular electrodes constructed from large pore size mesh with a projected cross sectional area of 130 cm^2 (5.45 mm mesh width, 0.8 mm wire thickness, Omnimesh, Belgium) were used in the first 74 days of operation (phase 1) for both cathode and anode. Subsequently, electrodes with smaller pore size mesh cut into rectangles with a projected cross sectional area of 360 cm^2 (size #36.6, Solana NV, Belgium) were used from day 74 until day 98 (phase 2) for both cathode and anode. A distance of approximately 15 mm separated the electrodes. During the experimental period, a Bv of 0.49 ±0.04 gCOD/L d and a E_{ap} of 2.0 ±0.1 V were applied.

2.3.3 Electrochemical measurements

The cell voltage of the MEC-septic tank was applied by a direct current (DC) power supply (PS3010, HQ-Power, Belgium). The current was measured by the voltage difference over a 1Ω resistor in the electrical circuit between the DC power supply and the cathode. The cell voltage, current production, and the cathode potential were recorded every 5 min with a data- acquisition unit (HP 34970A, Agilent, USA). The potential of the cathode and anode electrode were monitored with a Ag/AgCl reference electrode (model RE-5B, BASi, UK). The reference electrodes were verified regularly against a saturated calomel electrode (3 M KCl) (+244 mV versus SHE at 25°C). Hourly averages of the electrochemical parameters were used for further calculations.

2.4 Analytical procedures

Liquid and gas samples were taken daily or every second day from the reactors. Total solids (TS), volatile solids (VS), suspended solids (SS), total ammonia nitrogen (TAN), total Kjeldahl nitrogen (TKN) chemical oxygen demand (COD), total phosphorous (TP) and pH were determined according to Standard Methods [20]. Nitrite, nitrate and phosphate were determined with a 761 compact ion chromatograph equipped with a conductivity detector (Metrohm, Zofingen, Switzerland). Multi-elemental analyses (Fe, Ca, K, Mg, Na, S and Cu) were performed using an inductively coupled plasma optical emission spectrometer (ICP−OES, VarianVista MPX, USA). Volatile fatty acids (VFA) were, after extraction in diethyl ether, analyzed with a flame ionization detector (FID) gas chromatograph (GC-2014, Shimadzu). The lower detection limit for VFA analysis was 2 mg/L. Gas chromatography (CompactGC, Global Analyser Solutions, The Netherlands) with a thermal conductivity detector (TCD) was used for biogas analysis. Gas detection tubes (RAE systems Inc., USA) were used for the detection of hydrogen sulphide in the biogas with a detection limit of 1 ppmv.

3. Results

3.1 Reactor performance

Anaerobic digestion of black water under mesophilic septic tank conditions (control reactor) and in a MEC-septic tank was compared. Influent organic loading rates in both reactors were kept in the same order (0.49 ±0.04 gCOD/L d) throughout the whole study while the HRT varied (20-40 days) due to changes in the influent COD concentrations (Table 1). The MEC-septic tank was operated with two different electrode surface areas (130 cm^2 during phase 1 and 360 cm^2 during phase 2) while keeping all the operational parameters the same.

At the beginning of the experimental period (day 1 - 30), the biogas production rates in the control reactor averaged 0.07 $L_{biogas}/L_{reactor}$ d which corresponds to 30 ±9% of the maximum theoretical production based on the Bv applied (Fig. 2). The biogas production rates decreased from day 30 onwards stabilizing at only 0.01 $L_{biogas}/L_{reactor}$ d which is equivalent to 3.0 ±0.1% of the maximum theoretical production.

The biogas production rates in the MEC-septic tank during the first 25 days (phase 1) averaged 0.11 $L_{biogas}/L_{reactor}$ d, accounting for 44 ±5% of the maximum theoretical production (Fig. 2). Between day 25 and day 50 (phase 1), the biogas production rates were lower but stable at 0.07 $L_{biogas}/L_{reactor}$ d (32 ±4% of conversion). By the end of phase 1 (day 50-74), the biogas production rates slightly increased up to 0.09 $L_{biogas}/L_{reactor}$ d but subsequently leveled at 0.06 $L_{biogas}/L_{reactor}$ d. Both electrodes were replaced at this point due to the corrosion of the anode. During phase 2, the biogas production had an early increase up to 0.09 $L_{biogas}/L_{reactor}$ d but reached 0.05 $L_{biogas}/L_{reactor}$ d by the end of phase 2 (Fig. 2). The anode compartment accounted for 11 ±3% of the total biogas produced in the MEC-septic tank.

During the first 60 days of phase 1 and the end of phase 2 (from day 90 onwards), the effluent COD_{total} values of the MEC-septic tank were lower compared with those of the control reactor (Fig. 3a). However, during the end of phase 1 and the beginning of phase 2 (day 60-90), there was no clear difference in the effluent COD_{total} concentrations. Comparing the influent in both reactors, the COD_{total} removal was of the same order of magnitude, that is, about 85%, for both reactors. The removal of suspended solids was also in the same order, that is, 90% or more, for both reactors.

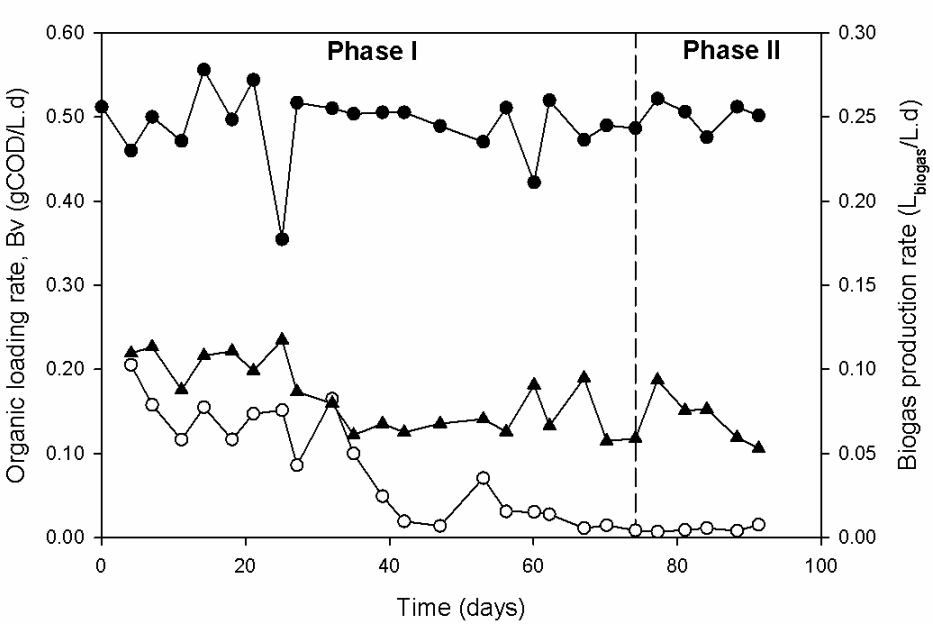


Figure 2. Performance of the lab-scale septic tank digester (control reactor) and the MEC-septic tank during the experimental period. (●) Organic loading rate in both reactors, Bv; (○) biogas production rate in the control reactor and (▲) biogas production rate in the MEC-septic tank.

The TAN values in the effluent of the MEC-septic tank were quite stable during phase 1 at an average of 630 ± 57 mgTAN/L (Fig. 3b). At the beginning of phase 2, TAN values increased up to 1 000 mgTAN/L but they subsequently decreased and stabilized to about 800 mgTAN/L. TAN values in the effluent of the control reactor had a decreasing trend from about 800 mgTAN/L (day 1) to approximately 600 mgTAN/L (day 60 onwards). From day 60 onwards, the TAN values in the effluent of the control reactor were stable and in the same order of magnitude as those of the influent (i.e. 600 mgTAN/L).

P_{total} values in the effluent of the MEC-septic tank were considerably lower than those of the control reactor (Fig. 3c). The P_{total} values of the MEC-septic tank averaged 77 ±14 mgPO_4-P/L during phase 1 and 34 ± 15 mgPO_4-P/L during phase 2 whereas the control reactor had a stable average of 112 ± 9 mgPO_4-P/L during the whole experimental period (39 ±22% lower P concentration than the control reactor). The P_{total} removed from the MEC-septic tank was approximately 60% in phase 1 and 89% in phase 2. The P_{total} removal in the control reactor was about 54%.

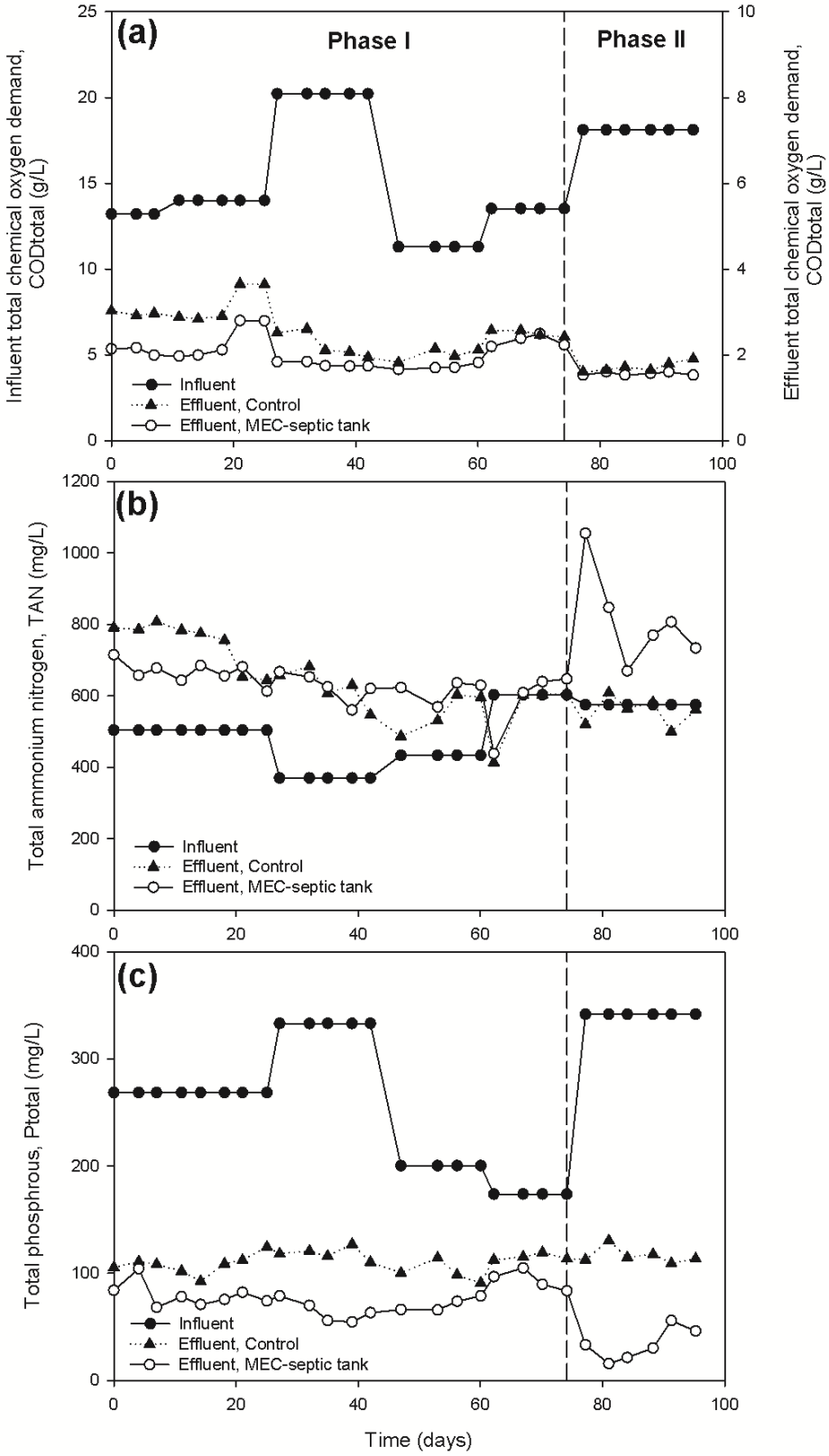


Figure 3. Evolution of the (●) influent and effluent concentrations in the (▲) control reactor and the (○) MEC-septic tank; (a) total chemical oxygen demand, CODtotal; (b) total ammonium nitrogen, TAN and (c) total phosphorous, Ptotal.

3.2 Chemical composition of electrode by-products

Under the conditions applied, electrochemical precipitation occurred on the surface of the cathode and anode sediments. The elemental chemical composition of the cathode precipitates during phase 1 on dry weight (DW) basis was 172 ± 7 mgP/gDW, 142 ± 9 mgMg/gDW), 65 ± 9 mgCa/gDW, 15 ± 2 mgK/gDW, 5 ± 1 mgNa/gDW), 1.4 ± 0.2 mgS/gDW, 15 ± 1 mgFe/gDW and to a lesser extent 0.03 ± 0.01 mgCu/gDW. The amount of precipitate in the cathode was in the order of 0.1 gDW/$cm^2_{projected\ area}$.

The composition of the anode sediments was 1416 mgP/gDW, 194 mgFe/gDW, 7 mgS/gDW, 13 mgCa/gDW, 4 mgCu/gDW, 3 mgMg/gDW, 4 mgNa/gDW and 3 mgK/gDW.

3.3 Biogas composition

The biogas composition with regard to methane (CH_4) and carbon dioxide (CO_2) remained fairly stable in all the compartments in both reactors during the experimental period. The biogas content in the MEC-septic tank was on an average 77 ± 3% of CH_4 and 23 ±3% of CO_2 in the cathode compartment and 68 ±2% of CH_4 and 32 ± 2% of CO_2 in the anode compartment. Low concentrations of H_2 (<2%) were detected only during the first 15 days of phase 1 in the cathode compartment. The CH_4 and CO_2 content in the control reactor were 71 ± 4% and 29 ± 4% and no H_2 was detected. Traces of hydrogen sulphide (H_2S) were detected in the biogas of the MEC-septic tank (cathode compartment) and of the control reactor (Fig. 4). Lower H_2S values in the MEC-septic tank were measured between day 25 to day 60 of phase 1 and during phase 2, they became below detection limits (<2 ppmv). The H_2S concentrations in the control reactor were as high as 700 ppmv (day 1-20) and on average about 200 ppmv (day 20-98).

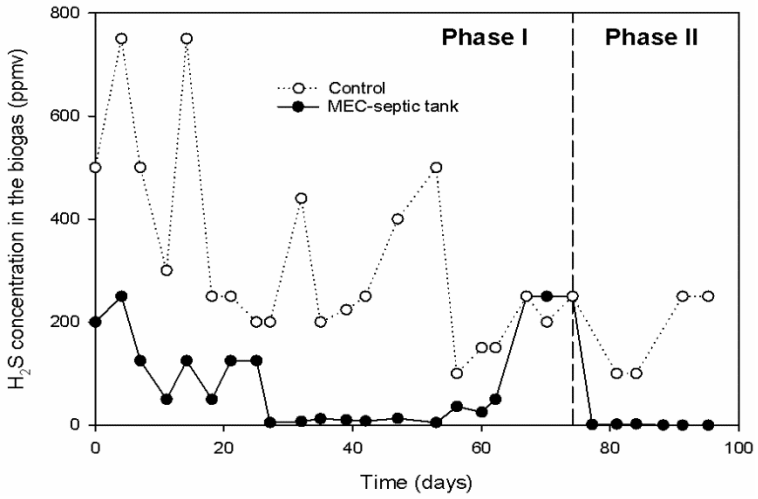


Figure 4. Hydrogen sulfide (H_2S) concentration in the biogas of (○) the standard septic tank digester (control reactor) and (●) the MEC-septic tank (cathode compartment) during the experimental period.

3.4 Current densities and electrode potentials in the MEC-septic tank

At an applied voltage of E_{ap} = 2.0 ±0.1V in continuous mode throughout the experimental period, current production was higher in phase 2 than in phase 1. During phase 1, the current in the MEC decreased from about 40 mA to an average of 20 mA corresponding to current densities of 3.5-1.9 A/m^2 (Fig. 5a). During the beginning of phase 2 (day 74-90), the current in the MEC reached a maximum value of approximately 180 mA (5 A/m^2 projected electrode area). From day 90 to day 98 (phase 2), the current stabilized at values around 40 mA (0.9 A/m^2 projected electrode area). The cathode potentials measured in the MEC were quite constant (Fig. 5b). During phase 1, cathode potential was on average -1.0 ± 0.2V versus SHE and on average -0.7 ± 0.01V versus SHE during phase 2. The anode potential was observed to reach a constant value in phase 1 of about +0.8 ± 0.2V versus SHE (Fig. 5c). During phase 2, the anode potential reached a higher continuous value of +1.2 ± 0.01V versus SHE. At stable operation, the internal resistance was estimated at 442 ± 273 Ω/m^2 (7.1 ± 0.9 mS/cm) in phase 1 and 105 ± 2 Ω/m^2 during phase 2 (5.0 ± 0.3 mS/cm). At these conditions, the power required to operate the MEC-septic tank can be estimated to be 90 ± 58 $Wh/m^3_{reactor}$ d.

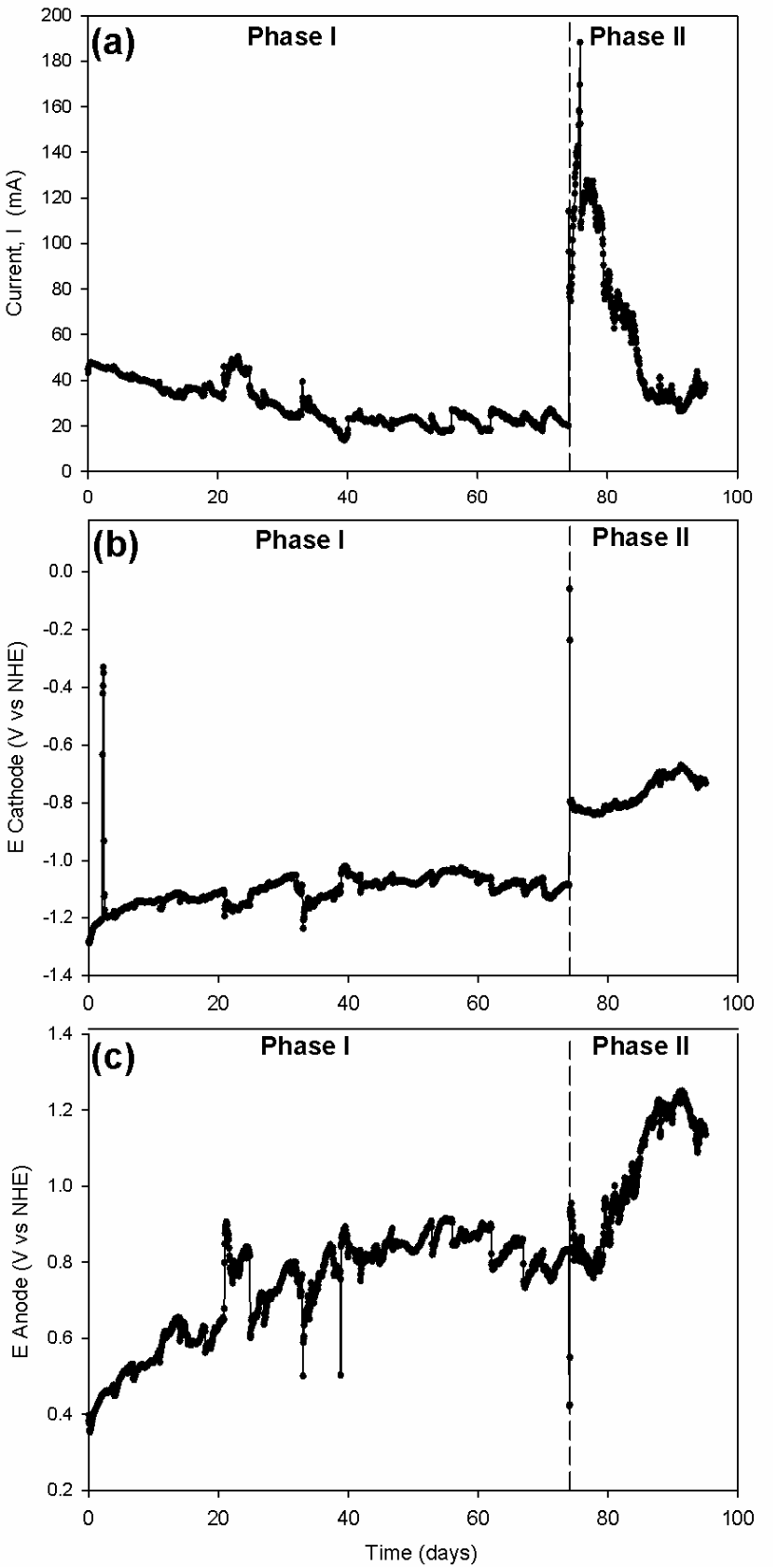


Figure 5. (a) Current versus time; (b) cathode potentials versus time and (c) anode potentials versus time. The MEC-septic tank was operated at an applied voltage of 2.0 ±0.1V.

4. Discussion

In this work, we investigated under lab-scale conditions the coupling of a MEC with a septic tank (MEC-septic tank) in terms of biogas production, biogas quality and effluent quality. To investigate the independent impact of the cathode and the anode on the biogas composition, a separation of the gas phase was installed in the reactor (Fig. 1). The liquid phase, however, did not have a physical separation or membrane installed; the cathode and anode compartment were in direct contact. The influent used was in terms of COD, nitrogen and phosphorous content comparable to black water as described by other authors i.e 7-38 gCOD/L, 0.9-1.5 gTN/L and 90-200 mgTP/L [21; 22].

The introduction of an electrochemical system into a septic tank had a positive effect on the overall performance and stability of the system, regardless of the electrode projected area. With a Bv of 0.49 ±0.04 gCOD/L d, an HRT between 20 and 40 days and an E_{ap} of 2.0 ± 0.1V (with stainless steel mesh as electrode material) under mesophilic conditions (30 °C), the COD conversion efficiency of the MEC-septic tank was stable at around 30% which was a factor of 5 higher than the control reactor. However, this performance is lower than in other septic tank-like reactors previously reported treating black water of similar composition as in this work. For instance, an UASB-septic tank operated with an HRT of 30 days was reported to have a COD conversion efficiency of about 60% at 25 °C and 39% at 15 °C, an accumulation system reached 58% COD conversion with an HRT of 150 days at 20 °C [6; 23]. The latter conversions related to systems which were in operation for long periods in which the biomethanation had fully developed; in our reactors, the time course of about 120 days corresponds to the start-up of a conventional septic tank in practice. Under the applied experimental conditions, our results suggest that the increase in methane production is partly due to H_2 generation in the cathode compartment. At the currents measured during operation of the MEC-septic tank, the rate of biogas (≈70% of CH_4) produced by electrochemically generated H_2 can be estimated to be a maximum theoretical of 0.007 $L_{biogas}/L_{reactor}$ d according to $V_{CH4} = (I \times t \times V_m)/(F \times \eta)$, where V_{CH4} is the volume of CH_4 produced, I is the current (A), t is the time (s), Vm is the molar volume of CH_4 (22.4 L/mol at STP), F is the Faraday constant (96 485 C/mol e^-) and η is the moles of electrons per mol of CH_4 ($4H_2 + CO_2 \rightarrow CH_4 + 2H_2O$, 8 moles e^-/mol CH_4). Hence, the direct contribution of the cathode by H_2 production is about 10% of the observed biogas production. One would also assume a contribution of the anode to increased hydrolysis due to the oxygen produced [10]; yet the latter compartment produced only some 10% of the biogas. Hence, the effect of the BES must be sought in an enhancement of the overall anaerobic conditions for the methanogens. Similar stimulatory effects have been reported before [18; 19]. Thus the installation of the MEC in the reactor strongly improved the stability of the conversion of organic matter to biogas (Fig. 2). This is

corroborated by the fact that during the steady operational period the amount of VFA in the reactor was below detection limit. In the control reactor, by contrast, the VFA varied in 156 ±238 mg/L.
It was expected that the biogas composition in the anode compartment would contain higher concentrations of CO_2 due to biological oxidation of COD. The CH_4/CO_2 composition in the anode compartment was similar as in the cathode compartment during the steady state operation. Yet, the H_2S in the anode biogas was below detection limit except when the anode was completely corroded reaching at this point maximum H_2S concentrations in the biogas up to 2 ppmv. However, the levels of H_2S in the cathode biogas compartment were also low (<50 ppmv), only reaching high levels (~250 ppmv) when the anode was corroded. One of the possible reasons might be that part of the sulphides diffuses toward the anodic compartment. This is in agreement with Rabaey et al. [24] who found that the diffusion of sulfides could occur even through a membrane. In addition, the corrosion of the anode (presence of iron salts in the bulk solution) could have a double positive effect on the H_2S removal. First, ferric and ferrous salts can chemically precipitate H_2S into iron sulphide salt. Second, metals can significantly increase the chemical sulfide oxidation [25]. The sacrificial anode will be dissolved in time due to the slow release of iron. Thus, it will require regular replacement. In this study, the anode was replaced after 90 days (considering the start-up period). Inexpensive materials such as cast iron could be used as anode, but this has to be further investigated.
The COD_{total} and the total suspended solid removal were in the same order of magnitude in both reactors (i.e. 85% and 90% respectively). This indicates that the COD removal was mainly due to the accumulation of suspended solids in both reactors. This is in agreement with Kujawa-Roeleveld et al. [6; 23] who reported COD removals of 78% and 61% for a UASB-septic tank operated at 25 °C and 15 °C respectively and 80% for an accumulation system run at 20 °C. The concentrations of TAN in the effluent of the MEC-septic tank during the first 20 days of phase 1 were unusually low considering that TAN values in the effluent should have been of the same order of magnitude than the organic anaerobic conversion due to protein biodegradation. The precipitation of TAN as struvite ($MgNH_4PO_4$) in the cathode compartments of MECs has previously been reported [26]. Indeed, it was observed that a thin layer of precipitates accumulated on the surface of the cathode due to electrochemical deposition (visual observation). The chemical composition of this precipitate suggested struvite crystallization (ratio P/Mg is close to 1). Not only P, Mg and N were found in the cathode precipitate but also Ca, K, Na, S, Fe and Cu. This indicated that the cathode could have nutrient recuperation capabilities because the precipitate could easily be separated by scraping it off from the surface. The productivity of the cathode precipitate could be estimated at about 12 $gDW/m^2_{electrode\ area}$ d. The electrochemical oxidation of iron in the anode has been reported to produce monomeric species (i.e. $Fe(OH)_3$) and polymeric hydroxy complexes which are also known as hydrous ferric oxides, HFO (i.e. $Fe(H_2O)_6^{3+}$, $Fe(H_2O)_5(OH)_2^{+}$, $Fe(H_2O)_4(OH)_2^{+}$, $Fe_2(H_2O)_8(OH)_2^{4+}$ and

$Fe_2(H_2O)_6(OH)_4^{4+}$ which are highly porous and have high surface areas with adsorption capabilities [27; 28]. We have also observed an amorphous sediment in the anode compartment that could be due to the formation of amorphous $Fe(OH)_3$ and/or HFO which is composed of mainly Fe but also of other compounds such as Cu, Mg, Ca, K, and Na that could be products of co-precipitation or adsorption. Other researches have shown that compounds such as Se, Ni, and As could be adsorbed by these hydroxides/polyhydroxides/polyhydroxyoxide metallic compounds [27-29]. Overall, our experiments indicate that nutrient recuperation was achieved, although the co-precipitation and adsorption mechanisms were not completely clear. Therefore, further research is warranted. Although most of the phosphorous removal was related to the removal of particulates, it was observed that the effluent of the MEC-septic tank contained a lower concentration of P_{total} than the control reactor (both with the same level of particulates, i.e ~0.5 gTSS/L). Phosphate might be precipitated by iron ions dissolved from the anode.

The energy required to operate the MEC-septic tank had a maximum average value of 86 $Wh/m^3_{reactor}$ d (5.2 A/m^2). These maximum energy consumptions were recorded in phase 2 where the electrodes projected area was larger (~3x) than in phase 1. Sasaki et al. [18] reported a 250 mL MEC reactor treating garbage slurry with a volumetric loading rate (Bv) of 31 gCOD/L d and an HRT of 45 days at thermophilic conditions (55 ºC) lower energy requirements and current densities at about 24 $Wh/m^3_{reactor}$.d (39 $\mu A/m^2$ projected electrode area and E_{ap} =1.73V). Tartakovsky et al. [10] reported a 3.5 L UASB-MEC with much higher power energy requirements and current densities; 4.8 $kWh/m^3_{reactor}$ d (75 A/m^2 and E_{ap} = 5.2 V) for a MEC treating garbage slurry with a Bv of 2-16 gCOD/L d and an HRT of 6-12 hours at mesophilic conditions (35 ºC). The amount of electricity that a MEC-septic tank could produce as biogas, subsequently converted by means of a co-generator system to electricity can be estimated at about 150 Wh/m^3 d (based on a Bv of 0.5 gCOD/Ld, 1 kg COD/kWh and a COD conversion efficiency of 30%). The energy required to run a MEC-system will consume roughly 60% of the net energy captured as biogas. From the energetic point of view, the system does not provide a great advantage. Yet, the MEC provides a higher quality for both the gaseous phase and the liquid effluents.

The MEC-septic tank could serve as an energy storage system. Sunlight energy could be directly incorporated in a septic tank by means of a photovoltaic (PV) cell without the need of transducer and battery system. For instance, the electricity cost of primary treatment of black water as carried out in our experiment is of the order of magnitude of € 5 per inhabitant equivalent per year (assuming a black water production of 45 L per inhabitant per day, a HRT of 30 days and an electricity cost of €0.1 /kWh). In our experience, about 40% of the electricity cost can be recovered as extra biogas. The difference of about € 3 per inhabitant equivalent per year represents the cost for the enhanced

treatment. These aspects warrant further and more long-term examination to properly define the applicability of the MEC-septic tank system for decentralized black water treatment.

5. Conclusions

In this study, a MEC was installed in a standard septic tank using stainless steel mesh as electrodes. In the MEC-septic tank during an operational period of 100 days and 20 days after start-up, a constant and stable biogas conversion efficiency of about 30% was achieved using black water as input. The system removed H_2S (77%) from the biogas and phosphorous (68%) from the liquid output. However, the system required a periodical replacement of the sacrificial anode. The electrical energy equivalents needed to implement the MEC system were estimated to be around 60% of the extra biogas captured. The overall concept of upgrading septic tanks by the installation of a MEC can be of particular interest in the context of improving the quality of the septic tank gaseous and liquids outputs, nutrient recuperation and with respect to temporary storage of small amounts of renewable energy in the form of biogas.

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